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Serial No.: 09/866,926  
Filed: May 30, 2001

Docket No.: MERCK-2264  
Applicant(s): HILARIUS et al.

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Date Filed: September 22, 2003

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IN THE UNITED STATES PATENT OFFICE

In the matter of an application for a German Patent in the name of Merck Patent GmbH, filed under No. 100 26 565.0 on 30 May 2000, and in the matter of an application for a United States Patent.

I, Dr. Ashwood Stephen DRANE, B.Sc., Ph.D., BDÜ, translator to Steve Drane Translations Ltd. of Beechwood, Chiverton, Hertfordshire, England, do solemnly and sincerely declare:

1. That I am a citizen of the United Kingdom of Great Britain and Northern Ireland.
2. That I am well acquainted with the German and English languages and am a competent translator thereof.
3. That the following is to the best of my knowledge and belief a true and correct translation of the above-referenced patent application and the Official Certificate attached thereto

Dated this 9th day of July 2003

Dr. Ashwood Stephen Drane

# FEDERAL REPUBLIC OF GERMANY



## Priority certificate regarding the filing of a patent application

**File reference:** 100 26 565.0

**Date of filing:** 30 May 2000

**Applicant/proprietor:** Merck Patent GmbH, Darmstadt/DE

**Title:** Ionic liquids

**IPC:** C 07 F, C 10 M, H 01 G

**The attached pages are a correct and accurate reproduction of the original  
documents of this patent application.**

**Seal**

Munich, 27 March 2001  
German Patent and Trademark Office  
On behalf of  
The President

[signature]

**Merck Patent Gesellschaft  
mit beschränkter Haftung**

**64271 D a r m s t a d t**

**Ionic liquids**

## Ionic liquids

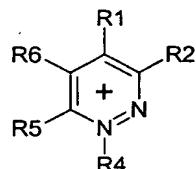
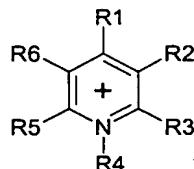
The invention relates to ionic liquids for use in electrochemical cells and organic syntheses.

- 5 Solvent-free ionic liquids or "salts which are molten at room temperature" were described for the first time in US 2446331. The problem with these strong Lewis acids is the formation of toxic gases on contact with atmospheric moisture.
- 10 Compounds involving  $\text{AlCl}_3$  and 1-ethyl-3-methylimidazolium (EMI) chloride have been investigated for a long time. Wilkes and Zaworotko presented novel solvent-free ionic liquids, EMI  $\text{BF}_4^-$  and EMI  $\text{O}_2\text{CCH}_3$ , in 1992 in J. Chem. Soc., Chem. Commun., p. 965. However, these compounds are unsuitable for use as electrolyte in 15 electrochemical cells since the  $\text{BF}_4^-$  and  $\text{CH}_3\text{C}_2^-$  anions are oxidised even at relatively low potentials.
- 20 WO 98/07729 describes a new class of conductive salts, the lithium borate complexes. These compounds have shown particularly good results in cycling experiments and have proven particularly stable. The borate salts are in the solid state and thus have relatively low conductivity.
- 25 US 5827602 describes the use of ionic liquids from the group consisting of pyridinium, pyridazinium, pyrimidinium, pyrazinium, imidazolium, pyrazolium, thiazolium, oxazolium and triazolium salts in electrochemical cells. These ionic liquids are particularly suitable for this application owing to good conductivities. The crucial disadvantage consists in the expensive synthesis of the raw materials, in particular the anions.
- 30 The object of the present invention is therefore to provide ionic liquids which have a large liquid range, high thermal stability and low corrosivity and anions which are less expensive to synthesise.
- 35 The object according to the invention is achieved by ionic liquids of the general formula

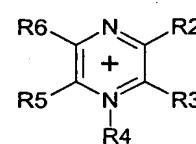
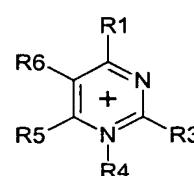
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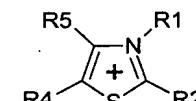
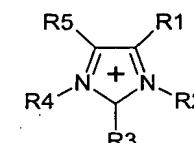
in which:

5  $K^+$  is a cation selected from the group consisting of

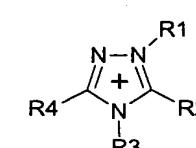
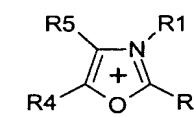
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15



20

25 where  $R^1$  to  $R^5$  are identical or different, are optionally bonded directly to one another by a single or double bond and each, individually or together, have the following meanings:

30 - H,

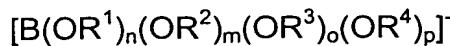
- halogen,

35 - an alkyl radical ( $C_1$  to  $C_8$ ), which may be partially or fully substituted by further groups, preferably F, Cl,  $N(C_nF_{(2n+1-x)}H_x)_2$ ,  $O(C_nF_{(2n+1-x)}H_x)$ ,  $SO_2(C_nF_{(2n+1-x)}H_x)$  or  $C_nF_{(2n+1-x)}H_x$  where  $1 < n < 6$  and  $0 < x \leq 13$ 

and

35  $A^-$  is an anion selected from the group consisting of

- 3 -



where  $0 \leq n, m, o, p \leq 4$ , and

5  $m+n+o+p=4$ ,

where  $R^1$  to  $R^4$  are different or are identical in pairs, are optionally bonded directly to one another by a single or double bond and are each, individually or together,

10 an aromatic ring from the group consisting of phenyl, naphthyl, anthracenyl and phenanthrenyl, which may be unsubstituted or monosubstituted or polysubstituted by  $C_nF_{(2n+1-x)}H_x$ , where  $1 < n < 6$  and  $0 < x \leq 13$ , or halogen (F, Cl or Br),

15 an aromatic heterocyclic ring from the group consisting of pyridyl, pyrazyl and pyrimidyl, which may be unsubstituted or monosubstituted or polysubstituted by  $C_nF_{(2n+1-x)}H_x$ , where  $1 < n < 6$  and  $0 < x \leq 13$ , or halogen or halogen (F, Cl or Br),

20 an alkyl radical ( $C_1$  to  $C_8$ ), which may be partially or fully substituted by further groups, preferably F, Cl, ,  $N(C_nF_{(2n+1-x)}H_x)_2$ ,  $O(C_nF_{(2n+1-x)}H_x)$ ,  $SO_2(C_nF_{(2n+1-x)}H_x)$ , or  $C_nF_{(2n+1-x)}H_x$ , where  $1 < n < 6$  and  $0 < x \leq 13$ ,

or  $OR^1$  to  $OR^4$ ,

25 individually or together, are an aromatic or aliphatic carboxyl, dicarboxyl, oxysulfonyl or oxycarbonyl radical, which may be partially or fully substituted by further groups, preferably F, Cl,  $N(C_nF_{(2n+1-x)}H_x)_2$ ,  $O(C_nF_{(2n+1-x)}H_x)$ ,  $SO_2(C_nF_{(2n+1-x)}H_x)$  or  $C_nF_{(2n+1-x)}H_x$ , where  $1 < n < 6$  and  $0 < x \leq 13$ .

30 These ionic liquids are suitable as solvents in organic synthesis, but also for use in electrochemical cells. In addition, the ionic liquids are suitable for use in the catalysis of chemical reactions. In addition, they can be used as inert solvents for highly reactive chemicals. A further area is use as hydraulic fluid.

It has been found that the compounds according to the invention are hydrophobic. Furthermore, the anhydrous synthesis minimises the undesired introduction of water into the system.

- 5 Surprisingly, it has been found that the ionic liquids do not corrode, but instead even passivate the aluminium current collector usually used in electrochemical cells. This enables the cycle stability to be increased. In addition, improved thermal stability of the system through the use of ionic liquids has been observed.
- 10 It has been found that the addition of solvents of low viscosity enables the conductivity to be improved. Low viscosity together with high conductivity is the prerequisite for use in electrochemical cells. The compounds according to the invention have a large liquid range, making them particularly suitable for these applications.
- 15 A prerequisite for use in double layer capacitors is high conductivity. The compounds according to the invention satisfy this criterion and can therefore be employed alone or in mixtures with other solvents or conductive salts. Suitable solvents are those selected from the group
- 20 consisting of organic carbonates (for example ethylene carbonate, propylene carbonate and derivatives thereof, butylene carbonate, dimethyl carbonate, diethyl carbonate, methyl ethyl carbonate, etc.), organic carboxylic acid esters (for example  $\gamma$ -butyrolactone, methyl formate, methyl acetate, ethyl acetate, ethyl propionate, methyl
- 25 propionate, methyl butyrate, ethyl butyrate, etc.), organic carboxylic acid amides (for example dimethylformamide; methylformamide, formamide, etc.), organic ethers (for example 1,2-dimethoxyethane, tetrahydrofuran, 2-methyltetrahydrofuran, tetrahydrofuran derivatives, 1,3-dioxolane, dioxane, dioxolane derivatives, etc.) or other aprotic
- 30 solvents (for example acetonitrile, sulfolane, dimethyl sulfoxide, nitromethane, phosphoric acid triesters, trimethoxymethane, 3-methyl-2-oxazolidinone, etc.). It is likewise possible to use solvent mixtures, such as, for example, ethylene carbonate/dimethyl carbonate (EC/DMC).

The compounds according to the invention can be used in electrolytes with conventional conductive salts. Examples of suitable electrolytes are those with conductive salts selected from the group consisting of LiPF<sub>6</sub>, LiBF<sub>4</sub>, LiClO<sub>4</sub>, LiAsF<sub>6</sub>, LiCF<sub>3</sub>SO<sub>3</sub>, LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub> and LiC(CF<sub>3</sub>SO<sub>2</sub>)<sub>3</sub>, and mixtures thereof. The electrolytes may also comprise organic isocyanates (DE 199 44 603) for reducing the water content. The electrolytes can likewise comprise organic alkali metal salts (DE 199 10 968) as additive. Suitable are alkali metal borates of the general formula

10



in which,

m and p are 0, 1, 2, 3 or 4 where m+p=4 and

R<sup>1</sup> and R<sup>2</sup> are identical or different,

15

are optionally bonded directly to one another by a single or double bond,

are each, individually or together, an aromatic or aliphatic carboxylic, dicarboxylic or sulfonic acid radical, or

20

are each, individually or together, an aromatic ring from the group consisting of phenyl, naphthyl, anthracenyl or phenanthrenyl, which may be unsubstituted or mono- to tetrasubstituted by A or Hal, or

25

are each, individually or together, a heterocyclic aromatic ring from the group consisting of pyridyl, pyrazyl or bipyridyl, which may be unsubstituted or mono- to trisubstituted by A or Hal, or

30

are each, individually or together, an aromatic hydroxy acid from the group consisting of aromatic hydroxycarboxylic acids and aromatic hydroxysulfonic acids, which may be unsubstituted or mono- to tetra-substituted by A or Hal,

and

35

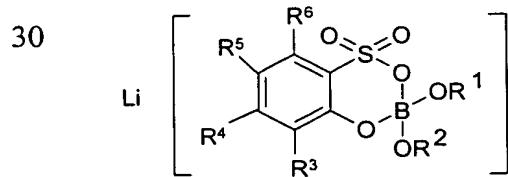
Hal is F, Cl or Br

and

A is alkyl having 1 to 6 carbon atoms, which may be mono- to trihalogenated. Likewise suitable are alkali metal alkoxides of the general formula

- 5                     $\text{Li}^+ \text{OR}^-$   
 in which R  
 is an aromatic or aliphatic carboxylic, dicarboxylic or sulfonic acid radical, or
- 10                  is an aromatic ring from the group consisting of phenyl, naphthyl, anthracenyl or phenanthrenyl, which may be unsubstituted or mono- to tetrasubstituted by A or Hal, or
- 15                  is a heterocyclic aromatic ring from the group consisting of pyridyl, pyrazyl and bipyridyl, which may be unsubstituted or mono- to trisubstituted by A or Hal, or
- 20                  is an aromatic hydroxy acid from the group consisting of aromatic hydroxycarboxylic acids and aromatic hydroxysulfonic acids, which may be unsubstituted or mono- to tetrasubstituted by A or Hal,  
 and  
 Hal            is F, Cl or Br  
 and
- 25                  A is alkyl having 1 to 6 carbon atoms, which may be mono- to trihalogenated.

Lithium complex salts of the formula



35                  where

5       $R^1$  and  $R^2$  are identical or different, are optionally bonded directly to one another via a single or double bond, and are each, individually or together, an aromatic ring from the group consisting of phenyl, naphthyl, anthracenyl or phenanthrenyl, which may be unsubstituted or mono- to hexasubstituted by alkyl (C<sub>1</sub> to C<sub>6</sub>), alkoxy groups (C<sub>1</sub> to C<sub>6</sub>) or halogen (F, Cl or Br),

10     or are each, individually or together, an aromatic heterocyclic ring from the group consisting of pyridyl, pyrazyl or pyrimidyl, which may be unsubstituted or mono- to tetrasubstituted by alkyl (C<sub>1</sub> to C<sub>6</sub>), alkoxy groups (C<sub>1</sub> to C<sub>6</sub>) or halogen (F, Cl or Br),

15     or are each, individually or together, an aromatic ring from the group consisting of hydroxybenzocarboxyl, hydroxynaphthalenecarboxyl, hydroxybenzosulfonyl and hydroxynaphthalenesulfonyl, which may be unsubstituted or mono- to tetrasubstituted by alkyl (C<sub>1</sub> to C<sub>6</sub>), alkoxy groups (C<sub>1</sub> to C<sub>6</sub>) or halogen (F, Cl or Br),

20     R<sup>3</sup> - R<sup>6</sup> may each, individually or in pairs and optionally bonded directly to one another by a single or double bond, have the following meanings:

1. alkyl (C<sub>1</sub> to C<sub>6</sub>), alkoxy (C<sub>1</sub> to C<sub>6</sub>) or halogen (F, Cl or Br)
2. an aromatic ring from the groups consisting of

25     phenyl, naphthyl, anthracenyl and phenanthrenyl, which may be unsubstituted or mono- to hexasubstituted by alkyl (C<sub>1</sub> to C<sub>6</sub>), alkoxy groups (C<sub>1</sub> to C<sub>6</sub>) or halogen (F, Cl or Br),

30     pyridyl, pyrazyl and pyrimidyl, which may be unsubstituted or mono- to tetrasubstituted by alkyl (C<sub>1</sub> to C<sub>6</sub>), alkoxy groups (C<sub>1</sub> to C<sub>6</sub>) or halogen (F, Cl or Br),

which are prepared by the following process (DE 199 32 317):

35     a) chlorosulfonic acid is added to 3-, 4-, 5- or 6-substituted phenol in a suitable solvent,

b) the intermediate from a) is reacted with chlorotrimethylsilane, and the reaction mixture is filtered and subjected to fractional distillation,

5 c) the intermediate from b) is reacted with lithium tetramethoxyborate(1-) in a suitable solvent, and the end product is isolated therefrom, may also be present in the electrolyte.

The electrolytes may likewise comprise compounds of the following formula (DE 199 41 566):

10  $[(R^1(CR^2R^3)_k]A_x)_yKt]^+ N(CF_3)_2$

where

Kt= N, P, As, Sb, S or Se

15 A= N, P, P(O), O, S, S(O), SO<sub>2</sub>, As, As(O), Sb or Sb(O)

20 R<sup>1</sup>, R<sup>2</sup> and R<sup>3</sup> are identical or different and are H, halogen, substituted and/or unsubstituted alkyl C<sub>n</sub>H<sub>2n+1</sub>, substituted and/or unsubstituted alkenyl having 1-18 carbon atoms and one or more double bonds, substituted and/or unsubstituted alkynyl having 1-18 carbon atoms and one or more triple bonds, substituted and/or unsubstituted cycloalkyl C<sub>m</sub>H<sub>2m-1</sub>, mono- or polysubstituted and/or unsubstituted phenyl, or substituted and/or unsubstituted heteroaryl,

25 A may be included in R<sup>1</sup>, R<sup>2</sup> and/or R<sup>3</sup> in various positions,

Kt may be included in a cyclic or heterocyclic ring,

the groups bonded to Kt may be identical or different,

30 where

n= 1-18

m= 3-7

35 k= 0 or 1-6

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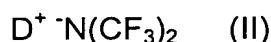
l= 1 or 2 in the case where x=1 and 1 in the case where x=0

x= 0 or 1

y= 1-4.

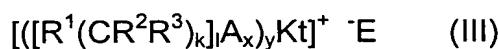
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The process for the preparation of these compounds is characterised in that an alkali metal salt of the general formula



10

where  $D^+$  is selected from the group consisting of the alkali metals, is reacted, in a polar organic solvent, with a salt of the general formula



15

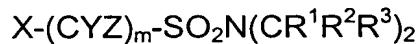
where

$Kt$ ,  $A$ ,  $R^1$ ,  $R^2$ ,  $R^3$ ,  $k$ ,  $l$ ,  $x$  and  $y$  are as defined above, and

$E$  is  $F^-$ ,  $Cl^-$ ,  $Br^-$ ,  $I^-$ ,  $BF_4^-$ ,  $ClO_4^-$ ,  $AsF_6^-$ ,  $SbF_6^-$  or  $PF_6^-$ .

20

However, use can also be made of electrolytes comprising compounds of the general formula (DE 199 53 638)



where

25

$X$  is  $H$ ,  $F$ ,  $Cl$ ,  $C_nF_{2n+1}$ ,  $C_nF_{2n-1}$  or  $(SO_2)_kN(CR^1R^2R^3)_2$

$Y$  is  $H$ ,  $F$  or  $Cl$

$Z$  is  $H$ ,  $F$  or  $Cl$

30

$R^1$ ,  $R^2$  and  $R^3$  are  $H$  and/or alkyl, fluoroalkyl or cycloalkyl

$m$  is 0-9 and, if  $X=H$ ,  $m\neq 0$

$n$  is 1-9

35

$k$  is 0 if  $m=0$  and  $k=1$  if  $m=1-9$ ,

prepared by reacting partially or perfluorinated alkylsulfonyl fluorides with dimethylamine in organic solvents, and complex salts of the general formula (DE 199 51 804)

5  $M^{x+} [EZ]_{xy}^{y-}$

in which:

x and y are 1, 2, 3, 4, 5 or 6

10  $M^{x+}$  is a metal ion

E is a Lewis acid selected from the group consisting of

15  $BR^1R^2R^3$ ,  $AIR^1R^2R^3$ ,  $PR^1R^2R^3R^4R^5$ ,  $AsR^1R^2R^3R^4R^5$  and  
 $VR^1R^2R^3R^4R^5$ ,

15  $R^1$  to  $R^5$  are identical or different, are optionally bonded directly to one another by a single or double bond, and each, individually or together, are

20 a halogen (F, Cl or Br),

an alkyl or alkoxy radical ( $C_1$  to  $C_8$ ), which may be partially or fully substituted by F, Cl or Br,

25 an aromatic ring, optionally bonded via oxygen, from the group consisting of phenyl, naphthyl, anthracenyl or phenanthrenyl, which may be unsubstituted or mono- to hexasubstituted by alkyl ( $C_1$  to  $C_8$ ) or F, Cl or Br,

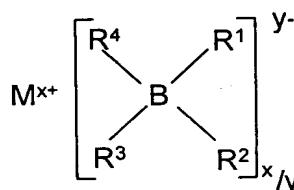
30 an aromatic heterocyclic ring, optionally bonded via oxygen, from the group consisting of pyridyl, pyrazyl or pyrimidyl, which may be unsubstituted or mono- to tetrasubstituted by alkyl ( $C_1$  to  $C_8$ ) or F, Cl or Br, and

35  $Z OR^6$ ,  $NR^6R^7$ ,  $CR^6R^7R^8$ ,  $OSO_2R^6$ ,  $N(SO_2R^6)(SO_2R^7)$ ,  
 $C(SO_2R^6)(SO_2R^7)(SO_2R^8)$ ,  
 $OCOR^6$ , where

$R^6$  to  $R^8$  are identical or different, are optionally bonded directly to one another by a single or double bond and are each, individually or together,

5 a hydrogen or as defined for R<sup>1</sup> to R<sup>5</sup>,  
prepared by reacting a corresponding boron or phosphorus Lewis  
acid/solvent adduct with a lithium or tetraalkylammonium imide,  
methanide or triflate.

## Borate salts (DE 199 59 722) of the general formula



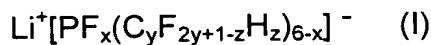
in which:

20. M is a metal ion or tetraalkylammonium ion,

x and y are 1, 2, 3, 4, 5 or 6,

25 R<sup>1</sup> to R<sup>4</sup> are identical or different and are alkoxy or carboxyl radicals (C<sub>1</sub>-C<sub>8</sub>), which are optionally bonded directly to one another by a single or double bond, may also be present. These borate salts are prepared by reacting lithium tetraalkoxyborate or a 1:1 mixture of lithium alkoxide with a borate with a suitable hydroxyl or carboxyl compound in a ratio of 2:1 or 4:1 in an aprotic solvent.

30 The compounds according to the invention may also be employed in electrolytes comprising lithium fluoroalkylphosphates of the general formula (I)



35 in which

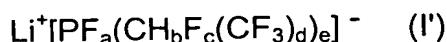
1 ≤ x ≤ 5

3 ≤ y ≤ 8

0 ≤ z ≤ 2y + 1

5

and the ligands ( $C_yF_{2y+1-z}H_z$ ) may be identical or different, with the exception of the compounds of the general formula (I')



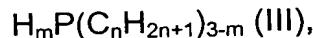
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in which a is an integer from 2 to 5, b = 0 or 1, c = 0 or 1, d = 2 and

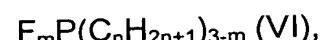
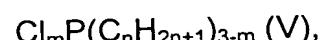
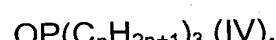
e is an integer from 1 to 4, with the provisos that b and c are not simultaneously each = 0, and the sum a + e is equal to 6, and the ligands ( $CH_bF_c(CF_3)_d$ ) may be identical or different (DE 100 089 55).

15

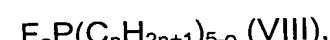
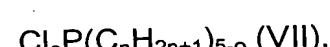
The process for the preparation of lithium fluoroalkylphosphates of the general formula (I) is characterised in that at least one compound of the general formula



20



25



in each of which

30

0 ≤ m ≤ 2, 3 ≤ n ≤ 8 and 0 ≤ o ≤ 4,

is fluorinated by electrolysis in hydrogen fluoride, the resultant mixture of fluorination products is separated by extraction, phase separation and/or distillation, and the resultant fluorinated alkylphosphorane is reacted with lithium fluoride in an aprotic solvent mixture with

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exclusion of moisture, and the resultant salt of the general formula (I) is purified and isolated by conventional methods.

5 The compounds according to the invention can be employed in electrolytes for electrochemical cells which contain positive-electrode material consisting of coated metal cores selected from the group consisting of Sb, Bi, Cd, In, Pb, Ga and tin or alloys thereof (DE 100 16 024). The process for the production of this positive-electrode material is characterised in that

10 a) a suspension or sol of the metal or alloy core in urotropin is prepared,

b) the suspension is emulsified with C<sub>5</sub>-C<sub>12</sub>-hydrocarbons,

15 c) the emulsion is precipitated onto the metal or alloy cores, and

d) the metal hydroxides or oxyhydrides are converted into the corresponding oxide by heat-treatment of the system.

20 The compounds according to the invention can also be employed in electrolytes for electrochemical cells having negative electrodes made from common lithium intercalation and insertion compounds, but also with negative-electrode materials consisting of lithium mixed oxide particles coated with one or more metal oxides (DE 199 22 522) by suspending the particles in an organic solvent, adding a solution of a hydrolysable metal compound and a hydrolysis solution to the suspension, and then filtering off, drying and optionally calcining the coated particles. They can also consist of lithium mixed oxide particles coated with one or more polymers (DE 199 46 066), obtained by a process in which the particles are suspended in a solvent, and the coated particles are subsequently filtered off, dried and optionally calcined. The compounds according to the invention may likewise be employed in systems having negative electrodes consisting of lithium mixed oxide particles with one or more coatings of alkali metal compounds and metal oxides (DE 100 14 884). The 30 process for the production of these materials is characterised in that the particles are suspended in an organic solvent, an alkali metal salt

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compound suspended in an organic solvent is added, metal oxides dissolved in an organic solvent are added, a hydrolysis solution is added to the suspension, and the coated particles are subsequently filtered off, dried and calcined.

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A general example of the invention is explained in greater detail below.

In order to prepare the anion selected from the group consisting of

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$[B(OR^1)_n(OR^2)_m(OR^3)_o(OR^4)_p]^-$

where  $0 \leq n, m, o, p \leq 4$ , and

$n+m+o+p=4$ ,

15

where  $R^1$  to  $R^4$  are different or are identical in pairs, are optionally bonded directly to one another by a single or double bond and are each, individually or together,

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an aromatic ring from the group consisting of phenyl, naphthyl, anthracenyl or phenanthrenyl, which may be unsubstituted or mono-substituted or polysubstituted by  $C_nF_{(2n+1-x)}H_x$ , where  $1 < n < 6$  and  $0 < x \leq 13$ , or halogen (F, Cl or Br),

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an aromatic heterocyclic ring from the group consisting of pyridyl, pyrazyl or pyrimidyl, which may be unsubstituted or monosubstituted or polysubstituted by  $C_nF_{(2n+1-x)}H_x$ , where  $1 < n < 6$  and  $0 < x \leq 13$ , or halogen or halogen (F, Cl or Br),

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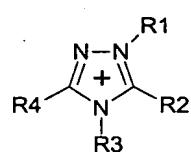
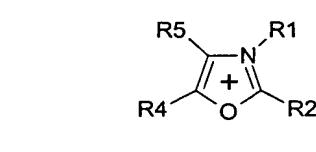
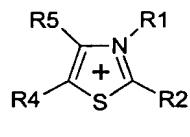
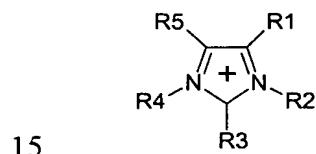
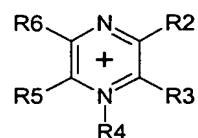
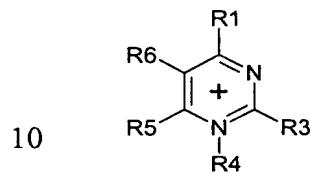
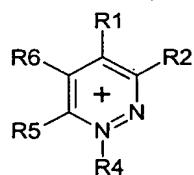
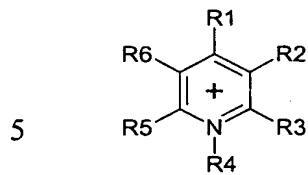
an alkyl radical ( $C_1$  to  $C_8$ ), which may be partially or fully substituted by further groups, preferably F, Cl, ,  $N(C_nF_{(2n+1-x)}H_x)_2$ ,  $O(C_nF_{(2n+1-x)}H_x)$ ,  $SO_2(C_nF_{(2n+1-x)}H_x)$  or  $C_nF_{(2n+1-x)}H_x$ , where  $1 < n < 6$  and  $0 < x \leq 13$ ,

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an aromatic or aliphatic carboxyl, dicarboxyl or hydroxycarbonyl radical, which may be partially or fully substituted by further groups, preferably F, Cl,  $N(C_nF_{(2n+1-x)}H_x)_2$ ,  $O(C_nF_{(2n+1-x)}H_x)$ ,  $SO_2(C_nF_{(2n+1-x)}H_x)$  or  $C_nF_{(2n+1-x)}H_x$ , where  $1 < n < 6$  and  $0 < x \leq 13$ ,

a known process from WO 98/07729 is used.

In order to prepare the cation selected from the group consisting of



20 a known process from US 5827602 is used. The starting materials are reacted for from about 0.5 to 12 hours, preferably 1-4 hours, in an aprotic organic solvent, at temperatures in the liquid range of the solvent.

25 In order to remove the by-products, the mixture is cooled to as far as -30°C, for example to from -10°C to -20°C in the case of LiCl as by-product, and the by-product which precipitates out is filtered off, preferably filtered off by vacuum.

30 The solvent/product mixture can be employed directly in the electrolyte. If desired, the solvent can also be distilled off and the resultant product dried.

The examples below are intended to explain the invention in greater detail, but without representing a limitation.

Examples

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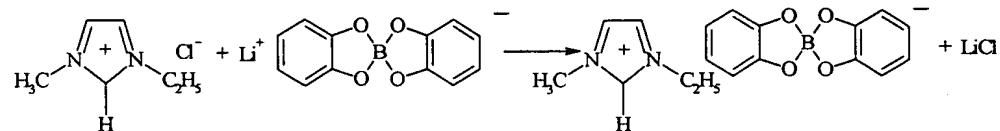
Example 1

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Synthesis of 1-ethyl-3-methylimidazolium bis[1,2-benzenediolato-O,O']borate

Lithium bis[1,2-benzenediolato-O,O']borate is synthesised in accordance with WO 94/27335 or WO 98/07729. The product is reacted in

15 acetonitrile in accordance with the following reaction equation:



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The reaction mixture is filtered by vacuum through a glass frit with cooling in order to remove the LiCl formed as by-product. The solvent is distilled off under reduced pressure, and the resultant 1-ethyl-3-methylimidazolium bis[1,2-benzenediolato-O,O']borate is dried under reduced pressure.

Example 2

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Synthesis of 1-ethyl-3-methylimidazolium bis[salicylato]borate

Lithium bis[salicylato]borate is synthesised in accordance with WO 94/27335 or in accordance with WO 98/07729 and reacted with 1-ethyl-2-methylimidazolium chloride in acetonitrile. The reaction mixture is filtered by vacuum through a glass frit with cooling in order to remove the LiCl formed as by-product. The solvent is distilled off under reduced pressure, and the resultant 1-ethyl-3-methylimidazolium bis[salicylato]borate is dried under reduced pressure.

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Example 3**Synthesis of 1-ethyl-3-methylimidazolium bis[oxalato]borate**

5       Lithium bis[oxalato]borate is synthesised in accordance with  
WO 94/27335 or in accordance with WO 98/07729 and reacted with  
1-ethyl-2-methylimidazolium chloride in acetonitrile. The reaction  
mixture is filtered by vacuum through a glass frit with cooling in order  
10      to remove the LiCl formed as by-product. The solvent is distilled off  
under reduced pressure, and the resultant 1-ethyl-3-methylimida-  
zolium bis[oxalato]borate is dried under reduced pressure.

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## Patent Claims

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## 1. Ionic liquids of the general formula

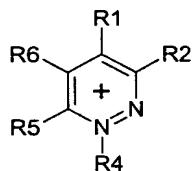
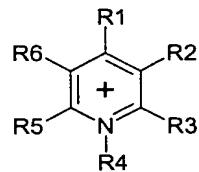


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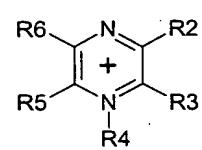
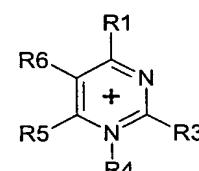
in which:

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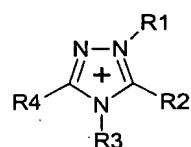
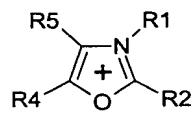
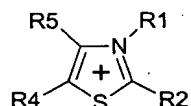
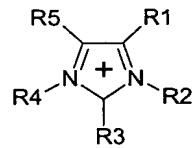
$K^+$  is a cation selected from the group consisting of



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where  $R^1$  to  $R^5$  are identical or different, are optionally bonded directly to one another by a single or double bond and each, individually or together, have the following meanings:

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- H,

- halogen,

- an alkyl radical (C<sub>1</sub> to C<sub>8</sub>), which may be partially or fully substituted by further groups, preferably F, Cl, N(C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>)<sub>2</sub>, O(C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>), SO<sub>2</sub>(C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>) or C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub> where 1<n<6 and 0<x≤13

5 and

A- is an anion selected from the group consisting of

[B(OR<sup>1</sup>)<sub>n</sub>(OR<sup>2</sup>)<sub>m</sub>(OR<sup>3</sup>)<sub>o</sub>(OR<sup>4</sup>)<sub>p</sub>]<sup>-</sup>

10 where 0≤n, m, o, p≤4, and

$$m+n+o+p=4,$$

15 where R<sup>1</sup> to R<sup>4</sup> are different or are identical in pairs, are optionally bonded directly to one another by a single or double bond and are each, individually or together,

20 an aromatic ring from the group consisting of phenyl, naphthyl, anthracenyl or phenanthrenyl, which may be unsubstituted or monosubstituted or polysubstituted by C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>, where 1<n<6 and 0<x≤13, or halogen (F, Cl or Br),

25 an aromatic heterocyclic ring from the group consisting of pyridyl, pyrazyl or pyrimidyl, which may be unsubstituted or monosubstituted or polysubstituted by C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>, where 1<n<6 and 0<x≤13, or halogen or halogen (F, Cl or Br),

30 an alkyl radical (C<sub>1</sub> to C<sub>8</sub>), which may be partially or fully substituted by further groups, preferably F, Cl, N(C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>)<sub>2</sub>, O(C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>), SO<sub>2</sub>(C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>) or C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>, where 1<n<6 and 0<x≤13,

35 or OR<sup>1</sup> to OR<sup>4</sup>,

35 individually or together, are an aromatic or aliphatic carboxyl, dicarboxyl, oxysulfonyl or oxycarbonyl radical, which may be partially or fully substituted by further groups, preferably F, Cl, N(C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>)<sub>2</sub>, O(C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>), SO<sub>2</sub>(C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>) or C<sub>n</sub>F<sub>(2n+1-x)</sub>H<sub>x</sub>, where 1<n<6 and 0<x≤13.

- 20 -

2. Use of ionic liquids as defined in Claim 1 in electrochemical cells having improved thermal stability, reduced corrosivity and a larger liquid range.

5 3. Use of ionic liquids as defined in Claim 1 in supercapacitors.

4. Use of ionic liquids as defined in Claim 1 as solvents and in the catalysis of chemical reactions.

5. Use of ionic liquids as defined in Claim 1 as hydraulic fluid.

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**Abstract**

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The invention relates to ionic liquids for use in electrochemical cells and for organic syntheses.

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